

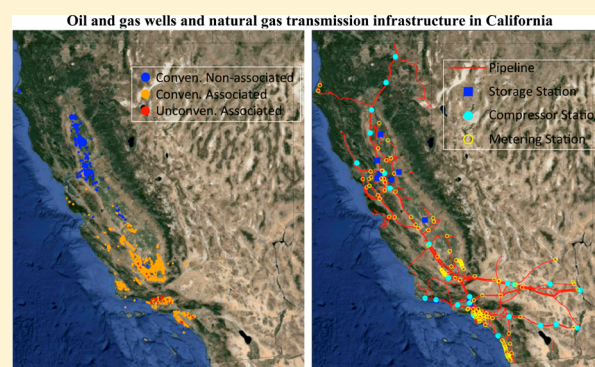
Spatially Explicit Methane Emissions from Petroleum Production and the Natural Gas System in California

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S Supporting Information

ABSTRACT: We present a new, spatially resolved inventory of methane (CH_4) emissions based on US-EPA emission factors and publically available activity data for 2010 California petroleum production and natural gas production, processing, transmission, and distribution. Compared to official California bottom-up inventories, our initial estimates are 3 to 7 times higher for the petroleum and natural gas production sectors but similar for the natural gas transmission and distribution sectors. Evidence from published “top-down” atmospheric measurement campaigns within Southern California supports our initial emission estimates from production and processing but indicates emission estimates from transmission and distribution are low by a factor of approximately 2. To provide emission maps with more accurate total emissions we scale the spatially resolved inventory by sector-specific results from a Southern California aircraft measurement campaign to all of California. Assuming uncertainties are determined by the uncertainties estimated in the top-down study, our estimated state total CH_4 emissions are $541 \pm 144 \text{ Gg yr}^{-1}$ (as compared with 210.7 Gg yr^{-1} in California’s current official inventory), where the majority of our reported uncertainty is derived from transmission and distribution. We note uncertainties relative to the mean for a given region are likely larger than that for the State total, emphasizing the need for additional measurements in undersampled regions.



1. INTRODUCTION

After 30 years of flat output, US natural gas withdrawals increased by 27% between 2006 and 2012¹ and are expected to increase further in the coming years.² While natural gas has the potential to reduce total greenhouse gas (GHG) emissions by displacing electrical power generation from coal, small emissions of methane (CH_4) during production and distribution could negate these benefits. For example, Alvarez et al.³ estimate leakage from well to power plant burner must remain below 3.2% for natural gas power generation to provide climate benefits relative to coal power generation over all time frames. The scientific community has not reached a consensus regarding CH_4 emissions from natural gas systems, and hence the climate benefits of natural gas remain uncertain. National-scale estimates of CH_4 emissions as a percentage of gas produced vary significantly. Burnham et al.⁴ estimate emission rates of 0.97–5.47% for conventional gas production and 0.71 to 5.23% for shale gas production, while Howarth et al.⁵ estimate relatively high emission rates 1.7 to 6% for conventional gas and 3.6% to 7.9% of shale gas. Allen et al.⁶ estimate emissions from the natural gas production sector to be 0.42% of total production.

In California, GHG emissions from natural gas and petroleum systems are influenced by federal and state policies and historical practices. California consumes more natural gas than any state but Texas⁷ but imports 85% of gas consumed.

California is currently the second largest oil producer (after Texas) and has potential for greater production from the Monterey Shale formation (estimated to contain 15 billion barrels of recoverable oil).⁸ Much of California’s oil production involves enhanced recovery with steam injection. Hydraulic fracturing was reported for a subset of recent wells. Regarding regulations, California’s oil and gas infrastructure is arguably subject to the most comprehensive emissions control regulations in the US. Though historically focusing on air quality control requirements, recent legislation to limit California’s contribution to global climate change (Assembly Bill 32) requires state total emissions be reduced to 1990 levels by 2020.

There remains significant uncertainty in statewide CH_4 emission estimates for natural gas and petroleum systems. A series of measurement campaigns in the South Coast Air Basin (SoCAB), including work by Wunch et al.,⁹ Hsu et al.,¹⁰ Townsend-Small et al.,¹¹ Wennberg et al.,¹² and Peischl et al.,¹³ suggest varying contributions to total CH_4 emissions from petroleum and natural gas activities. Additionally, these “top-down” measurement campaigns suggest higher CH_4 emissions

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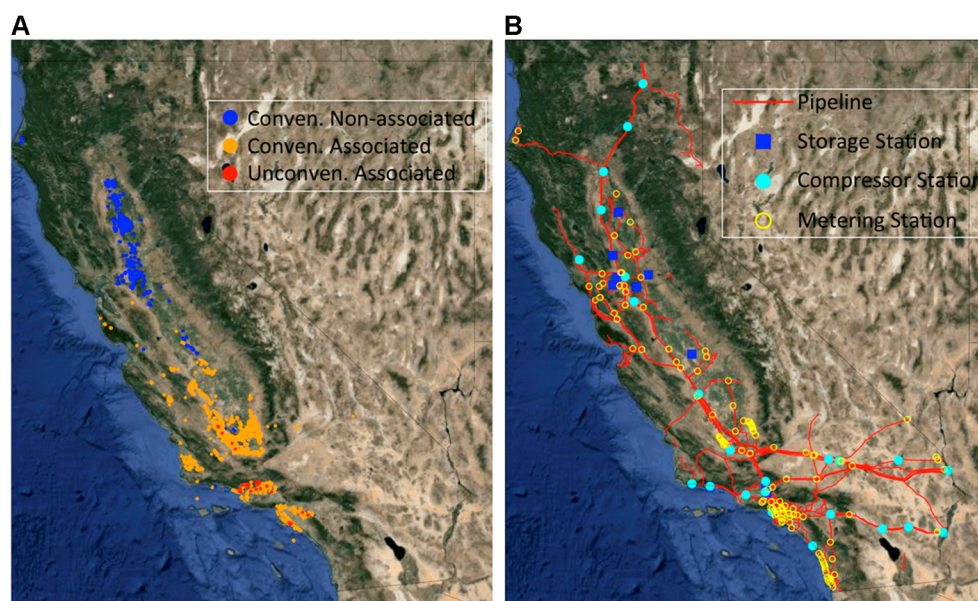


Figure 1. (A) Locations of individual oil and gas wells (active and new wells only) in California for the year 2010 (no unconventional nonassociated wells identified) and (B) map of the natural gas transmission system in California based on the CEC GIS database.

from petroleum and natural gas activities than is reported in the state's official GHG inventory¹⁴ or in a comprehensive California Air Resources Board (CARB) equipment survey of gas and petroleum producers.¹⁵ The discrepancy between "top-down" measurement campaigns and "bottom-up" CH₄ emission inventories is an important focus of this paper.

A spatially explicit CH₄ emission inventory is needed to compare state total oil and gas infrastructure CH₄ emission estimates with localized measurement campaigns. Additionally, a spatially explicit inventory can assist local planning estimates of GHG emissions and is an essential input to atmospheric inverse modeling techniques used to develop emission estimates from limited observations. The Emission Database for Global Atmospheric Research (EDGAR)¹⁶ includes emission estimates for natural gas and petroleum systems, but the spatial apportionment is primarily based on population.⁹ The California Greenhouse Gas Emission Measurements (CALGEM, calgem.lbl.gov) project inventory provides some refinement compared with the EDGAR inventory but was originally based on production data limited to northern California.¹⁷

The objectives of this paper are (1) to provide an updated spatially explicit bottom-up estimate of CH₄ emissions from the production, transmission, processing, and distribution of natural gas and petroleum production in California using spatially resolved activity information, disaggregated by sector and facility based on publicly available data and published emissions factors (e.g., US EPA^{18,19}) and (2) to characterize the implications of local measurement campaigns in Southern California on state total emission estimates.

The following Methods section details the data and emission factors used to develop the spatially explicit emissions inventory. The Results and Discussion section is subdivided to highlight estimated emission totals, compare this work to other bottom-up inventories, and then explore the differences between the bottom-up inventories and top-down measurement campaigns.

2. METHODS AND DATA

Overall Approach. We first estimate spatially explicit emissions for oil production and the natural gas system from four primary sectors: production, processing, transmission, and distribution. We then compare our emission estimates with existing statewide inventories and with top-down measurement campaigns in SoCAB. Finally, we scale state total emissions from our spatial inventory by sectors so that the SoCAB totals match top-down estimates by sector and estimate a range of revised state total emissions (see the Discussion section). Although we estimate uncertainty for our initial bottom-up CH₄ emissions, our primary goal is to estimate the uncertainty in the emission estimates by combining our spatially explicit emissions with measurement-based top-down analyses.

In general we attempt to disaggregate emissions to the finest scale provided by mostly publicly available activity data sets. For example, we estimate processing sector emissions based only on the volume of gas processed by each facility, while we estimate production sector emissions based on multiple factors including well type, workovers and liquid unloadings frequency, and other activity information. The sectors, subsectors, and associated activity data considered in this study are summarized in the Supporting Information (see Table S1).

We estimate emissions from the product of emission factors and activity data

$$E_X = E_f \times D_a \quad (1)$$

where E_X is the emission of species X (e.g., CH₄), E_f is the emission factor, and D_a is the associated measure of activity (e.g., annual volume of gas produced).

We then estimate grid emissions at a $0.1^\circ \times 0.1^\circ$ (roughly $10 \times 10 \text{ km}^2$) resolution as

$$E_X^p = \sum_{k=1}^n \sum_{j=1}^m \sum_{i=1}^l E_f^{i,j,k} \times D_a^{i,j,k} \quad (2)$$

where E_X^p is the emission of species X for grid cell p , $E_f^{i,j,k}$ is the emission factor for activity i , subsector j , and sector k with $\{i, j, k\}$.

$k\} \in p$, and D_a^{ijk} is the activity data for activity i , subsector j , and sector k . For example, for the pipeline subsector of the transmission sector, D_a^{ijk} represents the total length of pipelines within grid cell p . Therefore, given $\{i = \text{pipeline length}, j = \text{pipeline}, k = \text{transmission}\}$, D_a^{ijk} itself can be calculated as $D_a^{ijk} = \sum_{s=1}^{S_n} L_{s/p}$ where $L_{s/p}$ is the length of a pipeline segment belonging to grid cell p and S_n is the total number of pipeline segments in grid cell p . State total emissions for the entire system can be calculated as $\sum_{p=1}^T E_X^p$ where T is the total number of grid cells.

Data and Emission Factors. Production well activities are reported by the California Department of Conservation, Division of Oil, Gas, and Geothermal Resources (DOGGR).²⁰ DOGGR provides individual well locations, production volume for each well, well type, and other related information. Figure 1(A) shows the locations of individual natural gas wells (active and new) that produced natural gas. Figure 1(A) does not include the many abandoned or plugged wells. We adopt emission factors (see Tables S2 and S3 in the Supporting Information) from EPA for most activities.^{18,19}

We categorize wells in California into conventional and unconventional wells depending on the drilling/extraction method of gas or oil. In this study, unconventional wells are hydraulically fractured wells. The label “hydraulically fractured” in the DOGGR database²⁰ does not identify whether those wells were fractured vertically or horizontally (personal communication with DOGGR staff). Also, EPA attributes all emissions from dry gas production to conventional wells for the West Coast region, which includes California.^{18,19} We distinguish between associated gas production, from wells producing gas and petroleum, and nonassociated gas (dry gas) production, from wells producing only gas. We apply region-specific (i.e., West Coast) emission factors for nonassociated gas production available from EPA^{18,19} while we use national average emission factors for petroleum production (not available for individual regions).

The activity data gathered for the production sector varies by well type. For example, activity data for all active well types include production volumes of individual wells, well workover, and time of well completion, while liquid unloadings data are compiled only for dry gas wells (see Table S1). We find production of 187.7 billion cubic feet (Bcf) and 70.0 Bcf for associated and nonassociated natural gas, respectively, by summing across all individual wells within the California state.²⁰ These totals are slightly different from the DOGGR preliminary report, which estimated 184.6 and 70.9 Bcf, respectively.²¹ Associated production was mostly from conventional wells (186.5 Bcf), with 1.2 Bcf produced from unconventional wells. All nonassociated gas was produced from the conventional wells, and most was produced in Northern California (see Figure 1(A)).

Processing stage emissions are based on the volume of natural gas processed at each individual facility. Natural gas processing facility information is reported in the Natural Gas Annual Respondent Query System of Energy Information Administration (EIA).²² We provide processing facility locations in the Supporting Information (Figure S1). Because detailed component-level emission factors are not available for each facility, national average emissions factors used here are calculated based on the ratio of EPA’s processing sector emission estimates to total natural gas processed ($0.96 \text{ Tg CH}_4/308 \text{ Tg natural gas} = 3.1 \times 10^{-3} \text{ Mg CH}_4/\text{Mg natural gas}$; natural gas density of 19.05 g/cf used throughout the

study).^{18,22} The estimated emission factor is applied to the volume of natural gas processed at each individual facility.

Transmission emissions include emissions from pipelines, compressor stations, metering stations, and storage facilities, which are estimated using national average emission factors. We include the major transmission pipelines but not those used for local distribution of natural gas. Therefore, we assume that all other local pipes belong to the distribution sector, and this categorization may be different from that of EPA.

Georeferenced transmission sector data were obtained from the California Energy Commission (CEC) and are similar to available data from the CEC²³ and PG&E.^{24,25} Figure 1(B) shows spatial data for the transmission sector in California including pipelines, compressor stations, storage stations, and metering stations (distribution metering stations are not included). The total length of pipelines is 11.81×10^3 miles (excluding pipes marked with size of <0), slightly larger than EIA’s estimate of 11.77×10^3 miles.²⁶ Emission factors for transmission stage subsectors are summarized in the Supporting Information (see Table S4).

California compressor station locations are also included in the CEC GIS database (Figure 1(B)). We noted there were some unknown compressor stations listed in the CEC GIS database, and we visually checked and identified the unknown stations by referring to information from EIA²⁷ and PG&E.²⁸ Compressor station subsector emissions were estimated based on the simple emission factor of $571 \text{ Mg CH}_4/\text{station-year}$ from the Interstate Natural Gas Association of America (INGAA)²⁹ as information regarding subcomponents for each compressor station was not available (i.e., “Tier 1” approach).

Metering station and storage facility information is also available from the CEC GIS database and is included in Figure 1(B). We use an emission factor of $1.15 \text{ Mg CH}_4/\text{station-year}$ ²⁹ for identified metering stations. Identified storage facilities in the CEC GIS database are similar to storage facilities mapped by EIA³⁰ and PG&E.²⁵ Lacking detailed storage facility information (e.g., counts of pumps), we use an emission factor of $675 \text{ Mg CH}_4/\text{station-year}$ from INGAA.²⁹

Estimating emissions from the distribution of natural gas requires detailed geospatial information on distribution pipelines and metering stations. This information, especially detailed geospatial information, is not readily available. Therefore, we use the population density of California as a proxy for natural gas distribution. Population data was derived from the Center for International Earth Science Information Network (CIESIN) and Centro Internacional de Agricultura Tropical (CIAT)³¹ (see Figure S2). We note that there is a correlation between fractional county natural gas consumption and fractional county population (regression of consumption versus population = 1.07 ± 0.08 , $R^2 = 0.66$). We have not, however, adjusted our distribution estimates for large gas consumers such as power plants or industrial facilities as we have little knowledge of emissions from large gas consumers. We estimate a simple distribution emission rate of 0.3% of natural gas delivered to consumers by comparing the total natural consumption ($24.09 \times 10^{12} \text{ cf}$ for 2010³²) and the EPA-estimated CH_4 emissions for the natural gas distribution sector (1410 Gg CH_4 for 2010¹⁸). Similarly, a US EPA study estimated an emission rate (emission as percentage of produced or consumed gas) of 0.35% of the gross US natural gas production for the 1992 base year.³³ This emission rate is equivalent to 0.38% of the total US natural gas consumption for 1992 ($20.2 \times 10^{12} \text{ cf}$),³⁴ which is similar to our simple emission

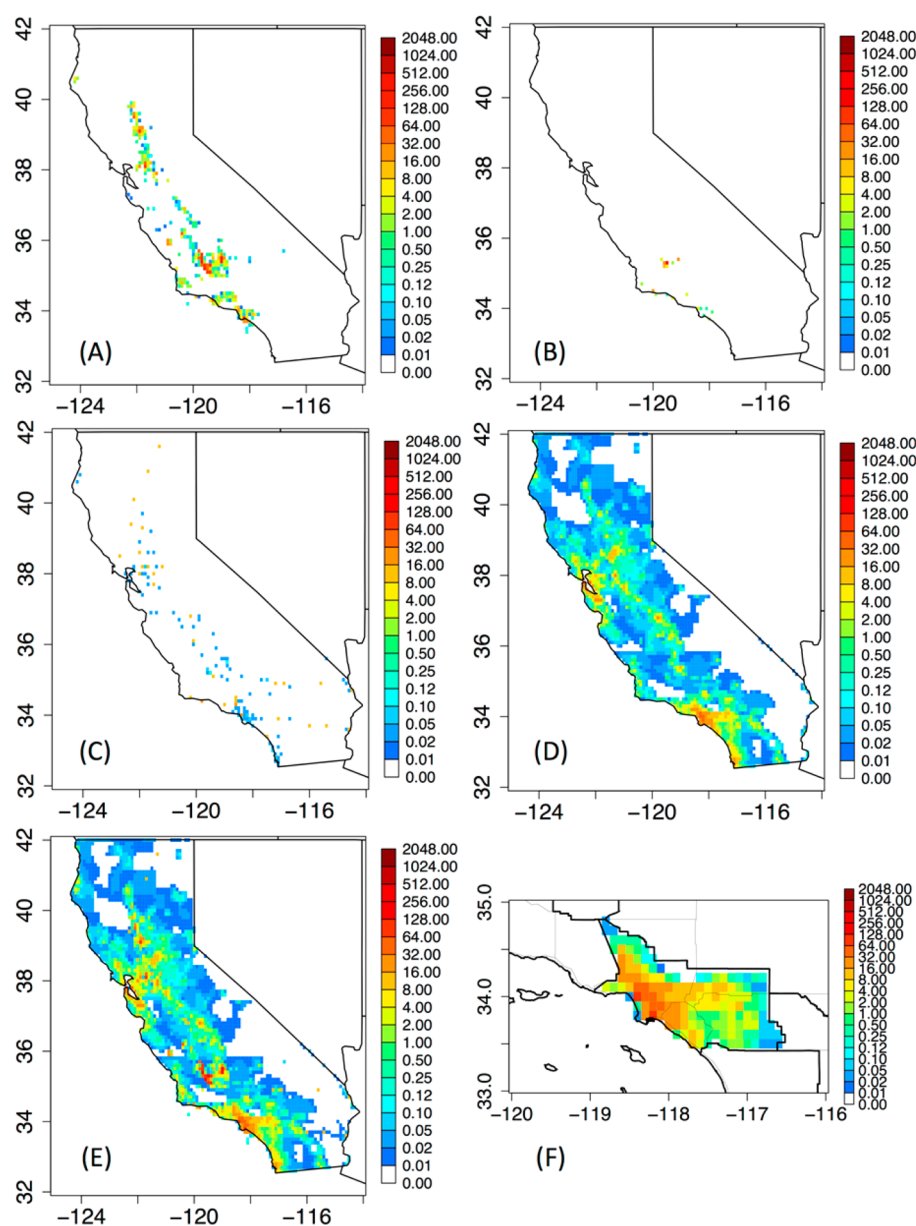


Figure 2. CH₄ emission maps (nmol m⁻² s⁻¹; 0.1° × 0.1°) for (A) production, (B) processing, (C) transmission, (D) distribution sectors in California, (E) state total CH₄ emissions, and (F) SoCAB total CH₄ emissions from petroleum production and the natural gas system. Emissions from federal offshore production are not included in the maps, but are calculated separately.

rate of 0.3%. Although we note that the emission rate for the distribution sector is highly uncertain, we use the national average emission rate of 0.3% in our initial estimation of distribution emissions. Later, we adjust the initial estimate based on results from a top-down analysis, which is California-specific. Also, a sensitivity analysis of emissions for the distribution sector to emission rates is presented in the Supporting Information. We apportion total CH₄ distribution emissions according to population density in California as in Jeong et al.,¹⁷ noting that this simplification does not capture variations in the age or maintenance characteristics of both utility-side and customer-side infrastructure.

3. RESULTS AND DISCUSSION

Initial Bottom-up Estimation of Spatially Explicit Emissions. We report spatially disaggregated CH₄ emission estimates for petroleum production and the production,

processing, transmission, and distribution sectors of the natural gas system. We construct emission maps (Figure 2) using the emission factors and activity data described in Section 2.

Figure 2(A) shows estimated CH₄ emissions from production based on well locations, well type, and other activity data. CH₄ emissions from nonassociated gas wells are dominant in Northern California (mostly Sacramento Valley), while CH₄ emissions from associated gas wells are concentrated in the southern San Joaquin Valley and SoCAB (see Figure 1(A)). We have not identified any unconventional non-associated gas wells from the DOGGR database. US EPA also reports that there is no unconventional non-associated gas well in the West Coast region.^{18,19} For oil wells (including associated gas production), we use emission factors for the petroleum system from EPA where there is no distinction in emission factors between conventional and unconventional wells.

Using the 2010 DOGGR database for oil and gas wells, we estimated total California CH₄ emissions (including offshore emissions) to be 140.2 and 27.7 Gg CH₄ yr⁻¹ for associated well and nonassociated well, respectively (see Figure S3 for emission maps for each well type). Although offshore petroleum production is 15% of California total production (nationally 29%), offshore oil well emissions account for 26% (offshore gas well emission = 1% of total gas well emission) of the total CH₄ emissions from oil wells.^{18,19,21,35} The reason for high CH₄ emissions from offshore petroleum production is that EPA estimates emission factors for the offshore petroleum production are 1.7 times higher than those of the onshore production.^{18,19}

Figure 2(B) shows CH₄ emissions from natural gas processing facilities. We estimated a state total of 12.10 Gg CH₄ yr⁻¹ for the processing sector or about 4% of the initial total CH₄ emission estimated in this study. Although total emissions from processing are relatively small, localized emissions are comparable to CH₄ emissions from small landfills in California.¹⁷

For the transmission sector, we estimate CH₄ emissions from major pipelines (local distribution pipelines are included in the distribution estimate), compressor stations, metering stations, and storage stations (Figure 2(C), see Table S4 for emission factors). CH₄ emissions for the pipeline subsector are estimated as a function of pipeline length, as described in Section 2. As size-dependent emission factors were unavailable we applied a uniform emission factor to all pipelines. We estimated annual state total CH₄ emissions from pipelines were 0.12 Gg CH₄, a fraction of the total transmission related emissions (20.90 Gg CH₄, see Figure S4 for pipeline emission map). Applying the emission factor from INGAA²⁹ to compressor stations uniformly, we estimated 13.14 Gg CH₄ emitted from compressor stations. Although less than 30 grid cells (0.1° × 0.1°) contained compressor stations, an accurate spatial characterization of these stations is important for research using top-down inverse modeling approaches. We estimated 0.14 and 7.50 Gg CH₄ for the metering station and storage facility subsectors, respectively. Among the four subsectors of the transmission sector, compressor stations account for the largest portion of the CH₄ emissions (63%) followed by storage facilities (36%) (see Figure S5 for emission maps). Pipelines and metering stations emit less than 2% of the transmission sector total.

We estimate state total emissions for the distribution sector are 130.0 Gg CH₄ yr⁻¹ assuming a 0.3% emission rate (ratio of CH₄ emissions to natural gas consumption, see Section 2). We apportion emissions in proportion to population density (see Figure 2(D)). Distribution accounts for 39% of the initial total CH₄ emission from oil production and the natural gas system in California, the large number driven by the large amount of natural gas that is consumed. We evaluate uncertainty in distribution (and other) sector emission estimates in relation to field measurement campaigns in the following sections.

Initial Bottom-up State Total Emissions. We estimate state total emissions from petroleum production and the natural gas system are 222–518 Gg CH₄ yr⁻¹ (see Table 1 and Figure 2(E)). The uncertainty estimate, shown in Table 1, is based on EPA's uncertainty estimates, which provides the overall uncertainty for the natural gas and petroleum systems as the percentage deviation from the mean estimates (see the Supporting Information for details). Later, however, we calculate adjusted emissions with uncertainties based on results

Table 1. Comparison of Methane Emissions Estimates (Gg CH₄ yr⁻¹) in California's Petroleum Production and Natural Gas System

| sector | initial bottom-up from this work (2010) | CARB-OGS (2007) ^a | CARB-GHG Inventory (2011) |
|---------------------------------|---|------------------------------|---------------------------|
| production: associated | 140.2 | 32.3 ^b | 25.1 ^c |
| production: nonassociated | 27.7 | 16.2 | -- |
| processing | 12.1 | 6.1 | 13.7 |
| transmission: storage | 7.5 | 6.3 | 4.2 |
| transmission: all other sources | 13.4 | -- | 167.7 ^d |
| distribution | 130.0 | -- | -- |
| total | 330.9 (222–518) ^e | 204.3 ^f | 210.7 |

^aSummary of CARB's Oil and Gas Industry Survey Results.¹⁵

^bIncludes all CH₄ emissions from associated gas and (on and offshore) crude oil production. ^cThe GHG inventory groups oil and gas extraction (associated + nonassociated) into one category. ^dThis includes the residential leakage and distribution pipelines as well as transmission pipelines because CARB used total organic gases emissions for specification of CH₄ emissions based on California Emission Inventory Development and Reporting System Database. ^eThis range of emissions (in the parentheses) was calculated by incorporating uncertainty estimates for the natural gas (lower bound = 19%, upper bound = 30% as percentage deviation from the mean at 95% confidence interval) and petroleum (lower bound = 24%, upper bound = 149%) systems from the EPA inventory,^{18,19} on which most of our emission factors are based. See the Supporting Information for details. ^fThis total is calculated by including the estimates from this work for the sectors where the CARB-OGS did not contain estimates.

from a top-down study in SoCAB and report them as our final estimates because we lack uncertainty estimates specific to California's activity data and emission factors.

We include analysis of sensitivity of emission estimates to selected factors, for example liquid unloadings and well completions, in the Supporting Information. CH₄ emissions at the production, processing, transmission, and distribution stages account for 50.7, 3.7, 6.3, and 39.3% of the initial state total emissions from petroleum production and the natural gas system (see Table 1 and Figure 2(E)).

We compare our initial bottom-up state total estimates to two other bottom-up estimates, the CARB Oil and Gas Survey (CARB-OGS)¹⁵ estimate and California's official greenhouse gas (CARB-GHG) inventory.¹⁴ Despite differences in methodology and in sector categorization, the bottom-up inventories are in rough agreement with each other except for emissions from petroleum production and, to a lesser extent, emissions from dry-gas production. The CARB-OGS was based on detailed information regarding equipment, equipment components and activity in oil and gas production and processing facilities in California. By gathering such detailed equipment information, CARB-OGS was able to apply detailed and activity-based emission factors and develop GHG emission estimates for individual facilities across the state. However, the specific facility data CARB-OGS gathered is confidential, and the reproduction of this type of bottom-up inventory would not be feasible for outside institutions.

Our estimate of total CH₄ emissions from petroleum (and associated gas) production is 4.3 times that of CARB-OGS. Our emission estimates from nonassociated gas production is 1.7 times that of CARB-OGS. Our estimate of total production

Table 2. Comparison of Natural Gas System CH₄ Emission Estimates in SoCAB (Gg CH₄ yr⁻¹)

| study | method | comments and sector specific totals | total emissions (all sources) | time period |
|-------------------------------------|---|---|--|-------------|
| Townsend-Small et al. ¹¹ | top-down: isotopic measurements | majority of all CH ₄ emissions from geologic seeps, pipelines, fossil fuels refining | | Aug. 2009 |
| Wunch et al. ⁹ | top-down: FTS, CH ₄ /CO ₂ and CH ₄ /CO correlation | two different estimates: relative to CO and CO ₂ | 400 ± 100 (CO-based) 600 ± 100 (CO ₂ -based) | 2007–2008 |
| Hsu et al. ¹⁰ | top-down: CH ₄ /CO correlation | NA | 380 ± 100 ^a | 2007–2008 |
| Wennberg et al. ¹² | top-down: CH ₄ /C ₂ H ₆ /CO correlation | natural gas (NG) system total: 390 ± 150 ^b | 440 ± 150 | 2008, 2010 |
| Peischl et al. ¹³ | top-down: CH ₄ /CO/CO ₂ correlation combined with light alkanes | NG transmission and distribution + local seeps: 192 ± 54 ^c landfills, dairies, wastewater: 182 ± 54 NG production/processing: 32 ± 7 | 410 ± 40 | 2010 |
| this work | bottom-up | NG production/processing: 24.0 ^d NG storage: 2.0 NG transmission and distribution ^e : 57.2 NG system total: 83.2 | NA | 2010 |
| CARB-GHG ¹⁴ | bottom-up ^f | NG production/processing: 2.5 NG storage: 1.1 NG transmission and distribution: 36.6 NG system total: 40.2 | NA | 2011 |
| CARB-OGS ¹⁵ | bottom-up ^g | NG production/processing/storage: 6.8 | NA | 2007 |
| EDGAR42 ¹⁶ | bottom-up | NG system total: 229.5 | 630 | 2008 |

^aOriginally Hsu et al.¹⁰ reported total LA County emissions (at 200 Gg CH₄ yr⁻¹), and Wennberg et al.¹² expanded the Hsu et al.¹⁰ results to the full SoCAB. ^bAn upper bound estimate. ^cPeischl et al.¹³ suggest an important portion of local seeps come from the La Brea Tar Pits as estimated by Farrell et al.³⁷ to be ~1/6 kiloton per day or ~61 Gg CH₄ yr⁻¹. This leaves 131 Gg CH₄ yr⁻¹ from transmission and distribution and seeps other than the La Brea Tar Pits. ^dThis includes CH₄ emissions from petroleum production. ^eDistribution estimated based on an initial assumption of 0.3% emission rate of total consumption. Storage not included in this line. ^fState totals scaled by spatial distribution from this work (as opposed to population as in past work). ^gSummarized from CARB's Oil and Gas Industry Survey Results, Final Report (Revised).¹⁵

emissions (petroleum based + dry natural gas) is 6.7 times larger than total production emissions from the CARB-GHG inventory, although the CARB-GHG production categories may not match exactly with our categories. Our estimates for emissions related to processing are double those of the CARB-OGS inventory, but our estimates for storage are within 20% of the CARB-OGS inventory. Our estimates of total transmission and distribution emissions are within 20% of CARB-GHG estimates (CARB-OGS did not estimate transmission and distribution emissions).

Comparison to Top-Down Estimates in SoCAB. Based on our initial bottom-up approach we estimate a total of 83.2 Gg CH₄ yr⁻¹ for the entire natural gas system and oil production in SoCAB (Table 2). By subsectors, we estimate 24.0 and 59.2 Gg CH₄ yr⁻¹ for natural gas production/processing and transmission/distribution, respectively. We compare our bottom-up CH₄ emission estimates in SoCAB (Figure 2(F)) to several top-down studies that have been conducted in the basin (Table 2). Comparison with these top-down studies allows us to evaluate the uncertainty in our bottom-up approach.

Recently, Wennberg et al.¹² estimated CH₄ emissions from the natural gas sector to be 390 ± 150 Gg CH₄ (Table 2) using atmospheric measurements of CH₄ and other trace gases. Wennberg et al.¹² used the ratio of C₂H₆ to CH₄ to estimate CH₄ emissions from natural gas, assuming natural gas systems are the only significant source of C₂H₆ in SoCAB. The Wennberg et al.¹² estimate is significantly larger (a factor of 4.7 ± 1.8) than our SoCAB estimate, suggesting our EPA-based emission factors applied in our initial bottom-up estimate are too low, at least for SoCAB. We note Shorter et al.³⁶ reported 1.6 ± 0.6% of total natural gas produced in the US was leaked during the production, processing, transport, and distribution of

the gas, roughly in-line with the 2% leakage estimate from Wennberg et al.¹²

Peischl et al.¹³ estimated separate CH₄ emissions in SoCAB for 1) production and processing (32 ± 7 Gg CH₄) and 2) natural gas transmission, distribution, and local seeps (192 ± 54 Gg CH₄), using measured ratios of alkanes (C₂–C₅) and published estimates for the alkane ratios of different sources in SoCAB. The total, 224 Gg CH₄, is lower than that of Wennberg et al.¹² Peischl et al.¹³ estimated a 17% CH₄ emission rate for natural gas produced in SoCAB. In explaining this high emission rate, they suggested that their estimate is within a factor of 1.5 of the CARB-OGS estimate. However, recent changes to the CARB-OGS¹⁵ have reduced estimated emissions from SoCAB production and processing so that they are lower than the estimates by Peischl et al.¹³ Our analysis suggests that one possible reason for this difference is in the relatively high offshore emissions. Our estimate, based on EPA emission estimates, for offshore production emissions in California (37.2 Gg CH₄ yr⁻¹) is 21 times higher than that of CARB-OGS, and 38% of the total is concentrated in SoCAB where associated gas production is dominant. Because we use different sets of emission factors for the natural gas and petroleum systems (effectively higher emission factors for associated gas produced with petroleum), we note that direct comparison of emissions with natural gas produced may not be appropriate for regions where associated gas production is significant. Another possible explanation for the high emission rate for natural gas production reported by Peischl et al. is that a large fraction of dry natural gas entering SoCAB is temporarily stored in depleted petroleum reservoirs where it has been found to be enriched in heavier alkanes (L. Sasadeusz, personal communication), such that leakage of stored gas may have an alkane profile similar to that locally produced associated natural gas. We note the volume of gas flow from storage wells is ~72 Bcf

for 2010 in SoCAB, compared to ~17 Bcf from production wells.^{20,21} If the production and processing category in Peischl et al.¹³ is redefined to include emissions from production, processing, and storage, then total emission rate is less than 3% of local associated gas production.

Assuming a similar alkane profile between stored and produced gas and including the storage sector with the production and processing sectors from Peischl et al.,¹³ we estimate total emissions from those sectors is 1.2 times our SoCAB inventory, while the emissions from transmission and distribution (not including storage or Tar Pit emissions, see Table 2) are 2.2 times greater in Peischl et al.¹³ than in our SoCAB inventory.

Adjusted State Emissions Based on SoCAB Measurements. Based on our discussion of the SoCAB field campaigns we make the general observation that the field campaigns find higher emissions (2–10 times larger depending on which inventories are compared) from natural gas systems and petroleum production than is indicated in the bottom-up inventories. In this section we utilize the spatially resolved bottom-up inventory developed here to ask the following question: if the leak rates implied by the SoCAB measurement campaigns are representative of the leak rates throughout the state, how much should we increase our estimates of total statewide emissions.

We choose to focus primarily on the top-down analysis in SoCAB by Peischl et al.¹³ because, although Wennberg et al.¹² estimate total emissions from the natural gas system, Peischl et al.¹³ attribute emissions separately to two different categories within the natural gas system: (1) the production/processing/storage sectors and (2) the transmission/distribution sectors. Note, as discussed above, we have chosen to group storage emissions with production and processing emissions based on the reported alkane profiles of storage gas. We also note that gas production in SoCAB is almost entirely associated with petroleum production, thus we do not make any changes to our bottom-up estimates of dry-gas production emissions.

A simple way to adjust our bottom-up estimate to be consistent with the measurements described by Peischl et al.¹³ is to scale our bottom-up emissions so the totals in SoCAB match the measurements. Here we multiply emissions from production, processing, and storage by 1.2 and emissions from transmission and distribution by 2.2. After this scaling, the inventory will reflect the application of the emission rates implied by Peischl et al.¹³ to the state total emissions.

Table 3 presents adjusted state-total emission estimates, where the total emissions within SoCAB are held equal to the emissions found by Peischl et al.¹³ For reference, the first column shows our initial bottom-up estimate based on EPA emission factors. The second column shows an even scaling (1.2 and 2.2, respectively) across sectors within the two categories: (1) emissions from the production, processing, and storage sectors and (2) emissions from transmission and distribution sectors. We also retain our initial estimates of emissions from dry-gas production.

We find an adjusted state total emission estimate of 541 ± 144 Gg yr⁻¹, where the uncertainty is derived solely from propagating uncertainties reported by Peischl et al.¹³ Transmission and distribution emissions represent 59% of the State total and also the majority of the uncertainty. Additional uncertainties are described below; however, we emphasize that uncertainties, relative to mean estimates in a given individual region of California, are likely larger than that for the State

Table 3. State Total CH₄ Emission Estimates (Gg CH₄ yr⁻¹) from California's Natural Gas System and Petroleum Production Constrained by Peischl et al.¹³ Measurements in SoCAB

| | EPA emission factor based bottom-up (this work) | bottom-up (this work) scaled to match measurement-based emission estimates |
|-------------------------------|---|--|
| Category 1^a | | |
| production (dry gas) | 28 ± 8 | 28 ± 8 |
| Category 2 | | |
| prod. associated | 140 | 172 |
| processing | 12 | 15 |
| storage | 8 | 9 |
| subtotal: | 160 | 196 ± 40 |
| Category 3^b | | |
| transmission | 13 | 30 |
| distribution | 130 | 288 |
| subtotal: | 143 | 317 ± 138 |
| total | 331 | 541 ± 144 |

^aCurrently there are no top-down estimates of dry gas emissions in California, so we leave the bottom-up estimates unchanged. The uncertainty in this category is based on EPA's uncertainty estimates for the natural gas system (an upper limit of 30%). ^bWe attempt to isolate emissions from transmission and distribution by subtracting from the Peischl et al.¹³ total major "local seep" emissions from the La Brea Tar Pits, (~1/6 kiloton per day) or ~61 Gg CH₄ yr⁻¹ as estimated by Farrell et al.³⁷

total. This highlights the necessity of making additional measurements in other regions.

There are two additional sources of uncertainty that we have not quantified. One source of uncertainty (see the Supporting Information and Table S5 for details) relates to which subsector is responsible for differences between the bottom-up and the top estimates. We note that sensitivity tests show variation in the adjusted state total of $\pm 7\%$ related to this source of uncertainty, which are within the uncertainty associated with the adjusted state total emission estimate. A second additional source of uncertainty is related to the degree to which emissions in SoCAB are representative of emissions across the state. We cannot definitively answer the question "how well do measurements in SoCAB represent emissions elsewhere in the State?" without additional top-down measurement campaigns or inverse modeling. To this end, California has begun top-down measurement campaigns and inverse modeling efforts. To date there have been only a limited number of studies focused on CH₄ emissions from the oil and gas industry in locations in California outside of SoCAB. In a recent example, Gentner et al. conclude, "the vast majority of CH₄ enhancements observed in the San Joaquin Valley (SJV) are due to emissions from dairy operations," as opposed to petroleum operations.³⁸ After adjusting our initial bottom-up estimates as described above, we estimate emissions in SJV from the production, processing, and storage sectors to equal 128.2 Gg yr⁻¹ and emissions from the transmission and distribution sectors of 34.4 Gg yr⁻¹, combined for a total 162.6 Gg yr⁻¹, 30% of our state total estimate. Jeong et al.³⁹ estimate emissions from dairy sources in SJV to be 1130 ± 205 Gg yr⁻¹, 7.0 times larger than our petroleum and natural gas estimate in SJV, in agreement with the qualitative conclusions by Gentner et al.³⁸

We also note that all the SoCAB measurement studies are consistent with the general conclusion that top-down estimates

indicate a higher rate of emissions than is assumed in bottom-up inventories, similar to results from recent top-down studies of air basins in Colorado by Pétron et al.⁴⁰ and in Utah by Karion et al.⁴¹

California reports 193.8 billion cf of associated gas production (onshore and offshore) in 2009 or 3.69 Tg yr⁻¹ (2009 data are used because federal offshore production is not included in 2010 data).⁴² We estimate 5.3 ± 1.1% (196 Gg/3.69 Tg) of this associated gas production total is leaked during associated production and all processing and storage phases of the natural gas system (see Table 3 for emission estimates). We can estimate leakage during the associated production phase only by subtracting the emissions we attribute to storage and processing to find an emission rate of 4.7% (172 Gg/3.69 Tg) for associated production only. However, we do not provide quantitative uncertainty for the leak rate estimate for associated production alone as we lack top-down estimates of emissions from the associated production sector alone.

Regarding dry gas, our bottom-up estimate indicates leakage of 1.8% of the DOGGR reported production (28 Gg yr⁻¹/1.58 Tg yr⁻¹).⁴² We note that our dry-gas estimate is based solely on the initial bottom-up estimate as opposed to our associated production emission estimate, which was adjusted to reflect the implications of the work by Peischl et al.

4. POLICY IMPLICATIONS

This paper makes three primary contributions to the policy discussion regarding CH₄ emissions from oil production and the natural gas system in California. (1) We present a spatially resolved CH₄ emission inventory based on detailed, publically available, facility and activity information. This inventory can be used for local and regional planning purposes as well as provide a key input for atmospheric inverse modeling. (2) We compare our bottom-up inventory to other bottom-up inventories developed by the State of California. Our bottom-up estimate of emissions from transmission and distribution is similar to those of the State. However, the CH₄ emission factors related to oil and gas production we apply lead to an emissions estimate 3–7 times larger than estimates by California. (3) We compare our bottom-up estimate to those from published measurement campaigns in California, primarily SoCAB (the air basin containing Los Angeles). The measurement campaigns to date support our higher emission estimates for the production sectors. However, the top-down analyses indicate that both our and California's bottom-up estimates for emissions from the transmission and distribution of natural gas are low by a factor of 2. Using our spatial inventory, we extrapolate emissions implied by a measurement study in SoCAB to generate new state total estimates of 541 ± 144 Gg yr⁻¹ (2.6 times the official California inventory), using the uncertainty provided in the measurement study, which likely represents a lower bound on uncertainty in total emissions. We note the importance of generating accurate CH₄ emission estimates as (at \$10 per metric ton CO₂ equivalent, and assuming 1-ton CH₄ is 33 tons CO₂ equivalent) 541 Gg yr⁻¹ is worth ~180 million dollars yr⁻¹.

Beyond the specific results relating to California, this work highlights the necessity for top-down analyses and inverse modeling to verify bottom-up estimates, as was the topic of a recent national level analysis.⁴³ We also have estimated emission rates from associated production, processing, and storage in California to be 4.7% and 1.8% for associated production and dry-gas production, respectively. Our analysis

indicates higher CH₄ emission rates from associated gas production (as opposed to dry-gas production). However, we argue that one might attribute CH₄ leakage from petroleum production to petroleum, as opposed to the relatively small amount of gas produced from those wells (~15% of the energy content produced from associated wells is natural gas, the rest is oil, see the Supporting Information for details). The argument for attributing these emissions to oil production is that oil is the primary product of oil wells with associated gas, while gas might be either flared or collected and sold as a secondary product. For example, in locations too remote to be affordably connected to natural gas infrastructure, oil is produced and associated natural gas is often simply flared. In those situations one would attribute the emissions from the produced and flared natural gas to the petroleum production. Why then attribute all CH₄ emissions from associated wells to natural gas if natural gas is sold rather than flared? It seems that some attribution of emissions between products is reasonable; we propose basing the attribution on the relative energy embedded in each product produced.

■ ASSOCIATED CONTENT

Supporting Information

Sector classification, emission factors used in this study, spatial locations of facilities, population density map in California, CH₄ emission maps for subsectors, and discussion of sensitivity sector emission factors. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

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